

## Highly time-resolved evolution of submicron aerosol chemical composition and optical properties during severe haze events in the largest megacity of China

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Despite of extensive efforts to characterization of the sources and formation mechanisms of severe haze pollution in eastern China, the accurate relationship of aerosol composition, mass-size distribution and optical properties during pollution episodes remain poorly understood. Here we conducted in-situ measurement on the mass size distribution of submicron aerosol (PM1) species by a High-Resolution Time-of-Flight Aerosol Mass spectrometer (HR-ToF-AMS), as well as particle light scattering by a Cavity Attenuated Phase Shift ALBedo monitor and Photoacoustic Extinctionmeter during the winter of 2017 in Shanghai, China. The average PM1 concentration was  $85.9 \pm 14.7 \mu\text{g}/\text{m}^3$  during the haze episodes,  $\sim 7$  times higher than that of clean period ( $12.1 \pm 3.1 \mu\text{g}/\text{m}^3$ ). Organic aerosol (OA) and inorganic species ( $\text{SO}_4^{2-} + \text{NO}_3^- + \text{NH}_4^+$ ) contributed 39.9% and 51.2% of the total mass of PM1 during the haze episodes, respectively. OA represented a single or bimodal distribution during the haze episodes with the peak concentration of  $51.8 \mu\text{g}/\text{m}^3$ . There were no obvious differences for ammonium nitrate and ammonium sulfate among the haze episodes, which represented single peak distributions at the size of 650 and 700 nm. and  $\sim 700$  nm, respectively. The peak position of OA, ammonium nitrate and ammonium sulfate in clean period were in the range of 450 - 500 nm, 550 - 600 nm and 450 - 500 nm with the peak concentration of  $5.5 \mu\text{g}/\text{m}^3$ ,  $3.1 \mu\text{g}/\text{m}^3$  and  $3.8 \mu\text{g}/\text{m}^3$ , respectively. Increased scattering coefficient in the haze episode was positively correlated with higher secondary inorganic aerosols and organic aerosol (OA). The high scattering coefficient contribution fraction peak diameter of ammonium nitrate and ammonium sulfate were in the range of 600 - 800 nm and 600 - 750 nm with the peak scattering coefficient of  $352.6 \text{ Mm}^{-1}$  and  $165.7 \text{ Mm}^{-1}$ . The size distribution of scattering for OA showed bimodal modes during all episodes. OA and ammonium nitrate were the largest contributor to scattering coefficients of PM1 during the haze episodes, accounting for 45.5% and 37.8%, respectively. The contribution of  $(\text{NH}_4)_2\text{SO}_4$  to the light scattering (24%) exceeded that of  $\text{NH}_4\text{NO}_3$  during clean period due to the enhanced sulfate concentrations. Our results elucidate substantial changes of optical properties due to chemical compositions and size distribution changes in the pollution period.